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**MIT JAR TESTS OF THE NATURAL POLYMER CHITOSAN
WITH FRESH POND WATER FROM THE
CAMBRIDGE WATER DEPARTMENT
NOVEMBER - DECEMBER, 1992**

Susan Murcott and Donald R. F. Harleman

MITSG 93-22

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Sea Grant College Program
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Cambridge, Massachusetts 02139

Grant No: NA90AA-D-SG424

Project No: 92-A-5

#4

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JANUARY, 1993

**by
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Funded by MIT Sea Grant

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1. INTRODUCTION

Cambridge Water Department (CWD), in common with many other water departments in the United States, uses the chemical alum as the primary coagulant in a multiple stage water treatment plant which is comprised of rapid mix, flocculation, clarification, and filtration units. For three decades, the resulting alum-based sludge has been discharged into Fresh Pond, which is also the City of Cambridge's terminal supply reservoir. This is not a long-term solids disposal option because Fresh Pond is filling up with sludge. Recent efforts to gain permission from the Massachusetts Water Resources Authority (MWRA) to discharge CWD solids into the sewerage system have not been well-received, due to MWRA restrictions on upstream discharge of metals (Cambridge Water Department, 1992).

Common sense as well as pollution prevention principles suggest that drinking water treatment chemicals should ideally be non-toxic, organic, biodegradable chemicals or inorganic substances that enhance sludge quality. Non-toxic chemicals will not contaminate drinking water. Organic and biodegradable chemicals will contribute to a beneficial, reusable sludge. The benefits and proven efficacy of aluminum salts in drinking water treatment are offset by the possible problems with residual metals contamination and the disposal of alum sludges. The growing popularity of synthetic organic polymers such as polyacrylamides and polyamines have the drawback of possible

toxicity problems. Toxicity concerns have led to a ban on the use of these polymers in drinking water treatment in Germany and the Netherlands (Jackson, 1992). Elsewhere, their dose is regulated to control potential problems with impurities (National Sanitation Foundation, 1990)

Today, organic polymers, whether natural or synthetic, are of interest in water treatment for the following reasons:

1. They are effective in very low dosages as compared to metal salts;
2. Low dosages of polymers reduce the volume of sludge produced;
3. Polymers improve the sludge dewatering process as compared with alum or iron salts;
4. Polymers are generally more biodegradable than alum or iron salt sludges and therefore ease sludge digestion by microorganisms;
5. They are noncorrosive and easy to handle;
6. Polymers have no problem with residual metals contamination.

The natural polymer examined in this study, chitosan, has some additional favorable characteristics:

1. It is an abundant and renewable resource;
2. It is biodegradable and has been shown to stimulate plant growth, making it a desirable component of a beneficial sludge;
3. It is non-toxic.

Natural organic polymers such as chitosan may provide an alternative to the use of alum or other inorganic metal salts in the coagulation process. In common with other chemical coagulants, the performance of chitosan can be evaluated through jar tests, an effective tool for comparing alternate chemical types, dosages and mixing regimes.

What is Chitosan?

Chitosan is a modified form of chitin, the second most abundant natural polymer after cellulose. Derived from chitin, found in the organic exoskeleton of crustacea such as crabs, shrimps, prawns and lobsters, chitosan is a polysaccharide, composed of poly-N-acetyl-glucoamine units, linked by beta 1-4 bonds into a linear polymer.

2. PURPOSE

The purpose of the MIT jar tests of chitosan using CWD water was to demonstrate the effectiveness of chitosan as a coagulant in drinking water applications. The approach was to compare the performance of the natural organic coagulant, chitosan, to the performance of alum and other chemical coagulants in terms of the parameters turbidity, color, pH and alkalinity. Twenty-five jar tests were conducted during November and December, 1992, at Parsons Laboratory, MIT, Cambridge, Massachusetts, on water sampled from the headworks to the Cambridge Water Department Treatment Plant.

3. PROCEDURE

Over the past four years, researchers at Parsons Laboratory working under Professor Donald Halleman, have gained extensive experience in jar testing and full-plant testing of a wide range of coagulants, coagulant aids, and flocculents. The first step in this process is to review existing practices at a given facility.

3.1 Mixing Regime

At the CWD laboratory, jar tests are conducted approximately every other week whenever an alum shipment is received. The purpose of the CWD jar tests is to verify the

viability of the material. Six jars are filled with raw water and alum is added to each jar in concentrations ranging from 15 to 40 mg/l. The alum is mixed rapidly for 2 minutes at 150 rpms, then the jars are stirred slowly for 30 minutes at 25 rpms. After 30 minutes of settling, a sample from each jar and also a raw water sample are analyzed for turbidity, color, and pH. This mixing regime is referred to in this report as the "CWD standard mixing procedure."

In the MIT jar tests, the CWD standard mixing procedure was followed in some tests and modified in others depending on whether alum, chitosan, or other primary coagulants and combinations of primary coagulants and coagulant aids were under investigation. Experiments were conducted with a variety of mixing times and speeds. Two basic mixing regimes were established:

- 1) The CWD standard mixing procedure;
- 2) A modification of the CWD standard mixing procedure based on increasing the time and speed of the rapid mixing step.

Although the modified mixing procedure did not improve alum's performance, it did show increased removal of turbidity and color with the chitosan/bentonite chemical regime. (Appendix A describes the 2 different mixing procedures).

3.2 Sampling

Water was sampled at the headworks to the CWD plant on various days during November and December, 1992. The sample was an 80:20 blend of Fresh Pond and Stony Brook Reservoir water, the blend typically used by the City of Cambridge. Fresh samples were collected in the morning of each test day in 5 gallon plastic buckets and transported to MIT.

3.3 Sample Analyses

MIT jar tests were conducted on a Phipps & Bird 6-paddle stirrer. Turbidity was analyzed using a HACH Model 2100P portable turbidimeter according to EPA Method 180.1 (nephelometric). Apparent color was analyzed using a HACH Model DR/2000 spectrophotometer according to the Platinum-Cobalt Standard Method (HACH Method 8025). This procedure assigns the wavelength of 455 nm as the dominant wavelength. pH was analyzed using a Cole-Palmer Digi-Sense LED #598-10.

4. RESULTS

4.1 Comparison of CWD and MIT Average Results

Figures 1, 2, and 3 show average alum jar testing results obtained in CWD tests versus MIT tests. These figures show that

there is reasonably good correlation between the 2 laboratory's sets of results in terms of turbidity removal, apparent color removal, and pH. The figures show that optimal removal of turbidity and color occurs at an alum concentration of about 20 to 30 mg/l. Also, as expected, the figures indicate that increased alum concentrations decrease the pH. The discrepancies between the 2 sets of average results can be explained by the use of different instruments for analysis and by the fact that MIT tests did not occur on site, i.e., immediately after sampling. MIT tests on a particular sample occurred over the course of a day, once water was sampled and transported back to Parsons Laboratory. Discrepancies in apparent color results (e.g. Figure 2a at an alum concentration of 15 mg/l) are also a function of the different dominant wavelengths used in the CWD standard procedure versus the MIT standard procedure. Whereas the CWD uses 401 nm as the dominant wavelength in their analytic method, MIT used 455 nm.

4.2 The Role of Bentonite

Chitosan alone performed poorly in terms of turbidity and color removal. However, in combination with bentonite, chitosan showed good results. Bentonite is a fine-grained inorganic clay of the mineral montmorillonite that assists in increasing the rate and efficiency of coagulation. It has a slight negative charge and can add weight to the flocs, joining them together and

producing larger, tougher flocs that will settle better. The clay particles are hydrated aluminosilicates of sodium, calcium, magnesium and iron. Clays are one of several basic types of commonly used coagulant aids in water treatment.

The initial tests using bentonite were to determine its optimal dose. Figures 4a and 4b show jar test results of 3 different concentrations of bentonite: 3, 6 and 9 mg/l, plus varying doses of chitosan. Both turbidity and color are best removed with a bentonite concentration of 9 mg/l. Bentonite concentrations in the range of 6 to 9 mg/l would be effective. Bentonite alone does not act as a coagulant and turbidity and color increased when bentonite was tested by itself. Bentonite did not improve the performance of alum.

4.3 Optimal Results with Chitosan Plus Bentonite

Having established an optimal bentonite concentration of 9 mg/l, the next step was to determine the optimal chitosan concentration. Figures 5 and 6 show that the optimal concentration of chitosan is about 0.5 mg/l. Figures 5a and 5b show turbidity concentration and turbidity percent removal respectively; Figures 6a and 6b show color concentration and color percent removal respectively.

4.4 Comparison of Optimal Results: Alum versus Chitosan/Bentonite

Alum was found to perform best when tested with the CWD standard mixing procedure. Chitosan was found to perform best when tested with the modified mixing procedure. Figures 7 and 8 compare the 2 different chemical regimes, alum or chitosan plus bentonite, based on their respective optimal mixing procedures. These figures illustrate the best results obtained on the same day on the same water sample with the mixing procedure selected to show the given chemical to its best advantage.

Turbidity concentration and percent removal using 0.5 chitosan plus 9 mg/l bentonite is an improvement over turbidity concentration and percent removal using 20 to 25 mg/l alum (Figure 7a and 7b). Color concentration and percent removal using 0.5 mg/l chitosan plus 9 mg/l bentonite is an improvement over color concentration and percent removal with 20 to 25 mg/l alum (Figure 8a and 8b). pH decreases slightly, by 0.1 to 0.2, in the optimal chitosan/bentonite sample relative to the raw water sample. pH reduction is more dramatic with alum addition, decreasing by a full 1.0 unit relative to the raw water sample. (Figure 9).

Some limited alkalinity results were also obtained. Those results indicate that alkalinity decreases by about 15% from 22 to 19 mg/l (as CaCO_3) over the range of effective chitosan/bentonite concentrations. Alkalinity decreases by over 50% from 30 to 14 mg/l (as CaCO_3) over the range of effective alum concentrations (Figure 10).

4.5 Other Chemical Coagulants Tested

Two other chemicals, ferric chloride (FeCl_3) and polyaluminum silico sulfate (PASS), were tested and compared to alum in a number of jar tests performed during November, 1991. Both chemicals performed comparably to alum at similar concentrations. Because, as an essential element in photosynthesis, iron may contribute to a beneficial sludge, ferric salts may be of interest for CWD. Ferric salts have been the metal salt of choice at some large water treatment facilities, such as the 70 mgd Hillsborough River Water Plant in Tampa, Florida (Matteo, J. 1992). The high cost of PASS was not outweighed by any obvious benefits based on the jar tests.

4.6 Chitosan as a Coagulant Aid

Chitosan was tested as a coagulant aid. In these tests, alum was used as the primary coagulant at 20 mg/l and chitosan was added as a coagulant aid in concentrations ranging from 0.2 to 2 mg/l. These tests did not show any advantage of using chitosan as a coagulant aid with alum. Alum alone performed better than alum plus chitosan.

5. COMPARISON OF MIT AND CDM JAR TEST RESULTS

Jar tests were a part of pilot studies performed by Camp Dresser and McKee Inc. at CWD during 1991 and 1992 (CDM, 1991a; CDM 1991b; CDM, 1992). Objectives of the pilot study included setting water quality goals of turbidity = 0.10 NTU and color = 5 cu (after coagulation and filtration) and minimizing the chemical dose and the overall amount of alum sludge produced. Table 1 lists the chemicals tested, their manufacturer and the type of polymer.

TABLE 1
CHEMICALS TESTED IN 1991, 1992 CDM PILOT STUDIES

Chemical	Manufacturer	Polymer Type
Alum	Holland	
Ferric Chloride		
Polyaluminum chloride (PAC)	Sternson, Inc.	
Magnifloc 572C, 573C, or 587C	American Cyanamid	cationic
Catfloc L	Calgon	cationic
Catfloc T-2	Calgon	cationic
Riverclear 101		
Magnifloc 1986N	American Cyanamid	nonionic
Magnifloc 985N	American Cyanamid	nonionic

Nine jar tests were conducted during the winter season, 1991, 8 on Fresh Pond water and 1 on Stony Brook water. Five jar tests were conducted during the summer season (1991) on the Fresh Pond/Stony Brook blend. Thirty-two jar tests were conducted

during the fall season, 1992, mostly on Stony Brook water. (Additional jar tests were conducted on Charles River water throughout the testing periods, but these will not be considered here.) Because all CDM jar test results are of water that has been filtered once or twice, they are not comparable to MIT jar test results, which have not been filtered. However, it is of interest to note the major CDM conclusions from these pilot tests as they pertain to the jar tests:

- 1) Cationic or nonionic polymers are required to assist primary coagulants. Effective concentrations range from 1 to 3 mg/l.
- 2) Alum, ferric chloride and polyaluminum chloride are effective primary coagulants. Effective concentrations range from 2 to 30 mg/l, depending on the source, water characteristics, and the particular primary coagulant.

Table 2 compares some of the CDM optimal jar test chemical chemicals and their concentrations with the MIT optimal jar test regime:

TABLE 2
COMPARISON OF CDM AND MIT OPTIMAL JAR TEST CHEMICALS AND DOSES

	Source	Primary Coagulant	Coagulant Aid
CDM (3/13/91)	Fresh Pond	Alum @ 10 mg/l	Magnifloc 573C @ 2 mg/l
CDM (4/8/91)	Stony Brook	PAC @ 6 mg/l	Magnifloc 573C @ 3 mg/l
CDM (8/9/91)	FP/SB Blend	FeCl ₃ @ 3 mg/l	Magnifloc 572C @ 2 mg/l
CDM (8/9/91)	FP/SB Blend	PAC @ 2 mg/l	Magnifloc 572C @ 2 mg/l
CDM (Fall, 92)	Stony Brook	Alum @ 30 mg/l	Magnifloc 572C @ 2 mg/l
CDM (Fall, 92)	Stony Brook	FeCl ₃ @ 20 mg/l	Magnifloc 572C @ 2 mg/l
MIT (12/21/92)	FP/SB Blend	Chitosan @ 0.5 mg/l	Bentonite @ 9 mg/l

6. SLUDGE PRODUCTION

A simple equation to evaluate sludge production is given as follows:

$$M_s = 86.4 Q(0.44A + SS + M) \quad (\text{Equation 1})$$

where

M_s = dry sludge produced (kg/day)
 Q = plant flow (m³/sec)
 A = alum dose (mg/l)
 SS = suspended solids in raw water (mg/l)
 M = miscellaneous chemical additions such as clay, polymers, etc.

The insoluble aluminum hydroxide complex $Al(H_2O)_3(OH)_3$ is

thought to predominate in most water treatment plant sludges. This species results in the production of 0.44 kg of chemical sludge for each kg of alum added. Any suspended solids present in the water will produce an equal amount of sludge. Polymers and clays will produce about one kg of sludge per kg of chemical addition (Davis, M, and Cornwell, D., 1985). Turbidity, especially in low turbidity waters such as at CWD, makes an insignificant contribution to sludge quantity.

Based on equation 1, a 25 mg/l concentration of alum will produce roughly the same sludge quantity as 0.5 mg/l chitosan plus 9 mg/l bentonite.

7. COST COMPARISON

A rough estimate can be made of the relative costs of using alum versus chitosan plus bentonite at the Cambridge Water Treatment Plant.

The following assumptions pertain:

Flow = 14 mgd (5.3×10^7 l/day)
Alum concentration = 25 mg/l
Chitosan concentration = 0.5 mg/l
Bentonite concentration = 9 mg/l
Cost of alum = \$0.10/lb (\$0.22/kg)
Cost of chitosan = \$3.00/lb (\$6.60/kg)¹
Cost of bentonite = \$0.22/kg (\$0.22/kg)

¹ Estimate provided by Lee Johnson, Vanson Chemicals, 7/92.

Alum Cost

$$\frac{25 \text{ mg}}{\text{L}} \times \frac{\$0.22}{\text{kg}} \times \frac{10^{-6} \text{ kg}}{\text{mg}} \times \frac{5.3 \times 10^7 \text{ L}}{\text{day}} = \frac{\$300}{\text{day}}$$

Chitosan + Bentonite Cost

Chitosan

$$\frac{0.5 \text{ mg}}{\text{L}} \times \frac{\$6.60}{\text{kg}} \times \frac{10^{-6} \text{ kg}}{\text{mg}} \times \frac{5.3 \times 10^7 \text{ L}}{\text{day}} = \frac{\$175}{\text{day}}$$

Bentonite

$$\frac{9 \text{ mg}}{\text{L}} \times \frac{\$0.22}{\text{kg}} \times \frac{10^{-6} \text{ kg}}{\text{mg}} \times \frac{5.3 \times 10^7 \text{ L}}{\text{day}} = \frac{\$103}{\text{day}}$$

$$\text{TOTAL} = \frac{\$278}{\text{day}}$$

In terms of the chemical cost alone, the chitosan/bentonite cost is slightly less expensive than the alum cost. However, besides the slight chemical cost advantage, the chitosan plus bentonite chemical combination would have other operating cost implications. Because chitosan plus bentonite has only a slight impact on pH, cost savings would occur from reduced demand for neutralizing agents. Because chitosan plus bentonite requires a higher mixing speed and mixing time, there would be some increased energy requirements. The most significant cost impact from the use of chitosan/bentonite could be in the savings in sludge handling and disposal. The increased options for disposal of beneficial sludge may outweigh the additional chemical and energy costs. Table 3, which gives internal estimates used by the Massachusetts Water Resources Authority (MWRA) for the evaluation

of various sludge handling and disposal alternatives (Schiemann, C. and Outwater, A., 1992), illustrates the wide range of sludge handling and disposal costs. The better the sludge, the greater the number of sludge disposal options available. This is an important subject which requires further investigation.

TABLE 3
COST ESTIMATES OF SLUDGE HANDLING AND DISPOSAL
(\$/dry ton of sludge)

	Processing	Transportation	Disposal	Total
Land Application	0	150	0	150
Compost	375	100	-10	465
Pellets	450	50	-50	450
Landfill	0	100	300	400
Chemfix	200	100	0	300
Incineration	300	100	0	400
Ocean Disposal	0	50	0	50

8. CONCLUSIONS

1. Low doses of the natural polymer, chitosan, plus bentonite are able to perform as well or better than alum in the removal of turbidity and color in jar tests of Cambridge Water Department drinking water from Fresh Pond.

2. In the range of optimal doses, alum reduces pH by 1.0 unit; chitosan plus bentonite lowers pH by 0.1 to 0.2 units. Water

treated with chitosan/bentonite will require less subsequent chemical treatment to neutralize the water.

3. The Cambridge Water Department standard mixing procedure showed alum to its best advantage. A modified mixing regime using faster mixing speeds and a longer mixing time optimized the performance of chitosan plus bentonite.

4. Limited alkalinity testing indicates that in the range of optimal doses, alkalinity is reduced by 15% with chitosan plus bentonite and by over 50% with alum.

5. Testing of other chemicals showed that ferric chloride or polyaluminum silica sulfate could obtain comparable turbidity and color removal to alum.

6. A 25 mg/l concentration of alum will produce roughly the same sludge quantity as 0.5 mg/l chitosan plus 9 mg/l bentonite.

7. The chemical cost of chitosan plus bentonite would be somewhat more expensive than alum at the current dose. Chemical costs alone do not give the complete picture. Effects of chitosan plus bentonite on the reduced need for pH adjustment chemicals, energy requirements, and sludge disposal options also need to be considered in computing the overall operating costs.

9. FURTHER STUDY

Chitosan plus bentonite appears to hold promise in the treatment of City of Cambridge drinking water. It performs favorably in jar tests and is a non-toxic chemical regime that would produce a beneficial sludge. Further study could correlate MIT jar test procedures with CDM jar test procedures in order to compare results. Tests on the effect of chitosan/bentonite on organics and inorganics, including TOC, DOC, THMs, TOXs, and metals would be undertaken. Chitosan/bentonite could also be tested in the pilot system currently set up at CWD. Detailed estimates of sludge production and overall costs should also be ascertained, based on data provided by CWD. A proposal for such a study is forth-coming.

10. REFERENCES

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APPENDIX A MIXING PROCEDURES

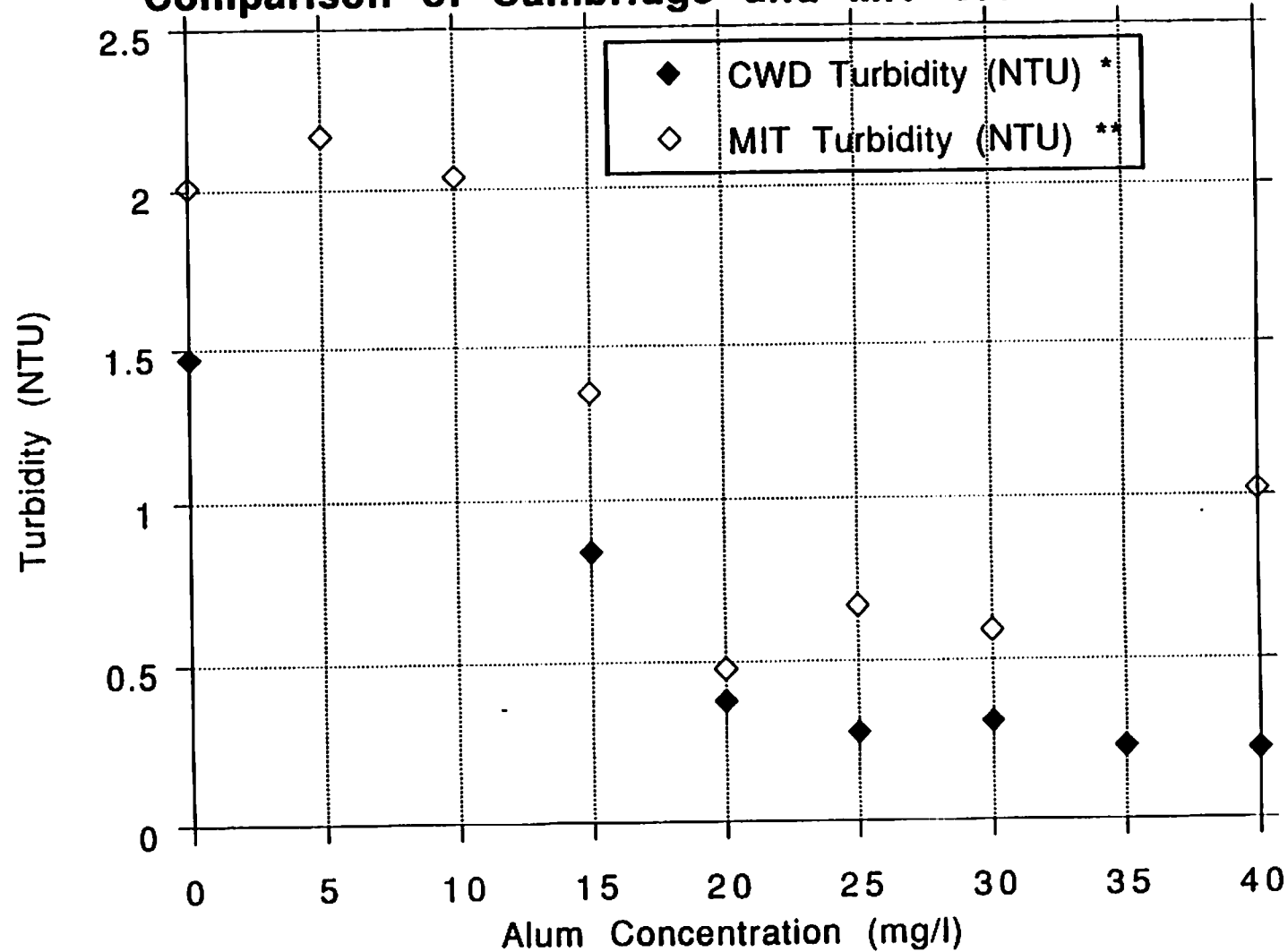
Cambridge Water Department Standard Mixing Procedure

1. Fill each of the 6 beakers with raw water.
2. Add alum to each beaker in increments of 5 mg/l, beginning with 15 mg/l.
3. Mix at 150 rpm for 2 minutes
4. Mix at 25 rpm for 30 minutes
5. Turn the mixer off and let settle for 30 minutes.

Modified Standard Mixing Procedure

1. Fill each of the 6 beakers with raw water.
2. Add bentonite.
3. Mix at 150 rpm for 1 minute
4. Add chitosan.
5. Mix at 300 rpm for 4 minutes
6. Mix at 150 rpm for 5 minutes
7. Mix at 50 rpm for 20 minutes.
5. Turn the mixer off and let settle for 30 minutes.

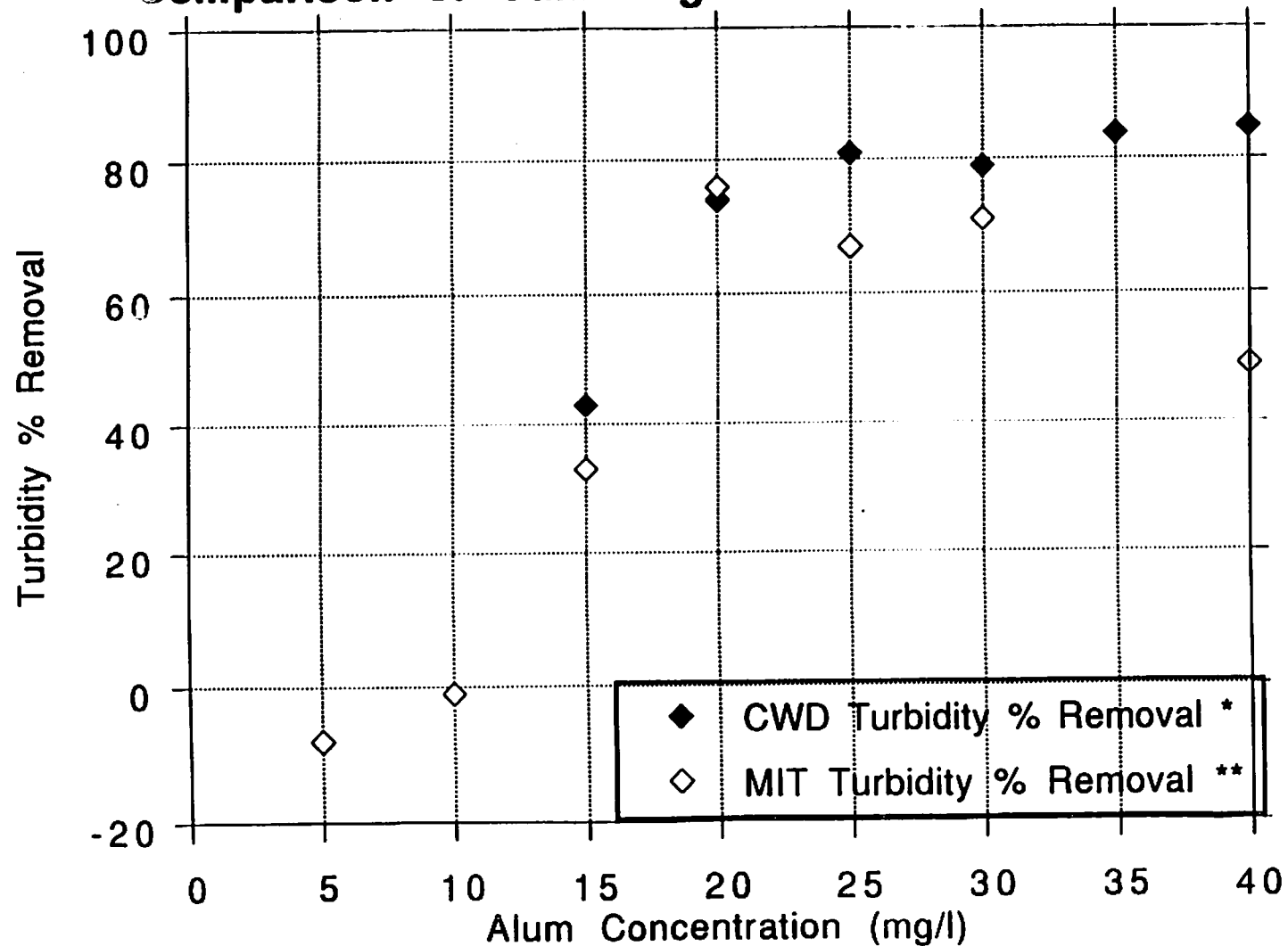
Figure 1a
Turbidity Concentration with Alum
Comparison of Cambridge and MIT Jar Test Results



* Average jar test data for Oct., Nov., and Dec., 1992

** Average jar test data for Nov. and Dec., 1992

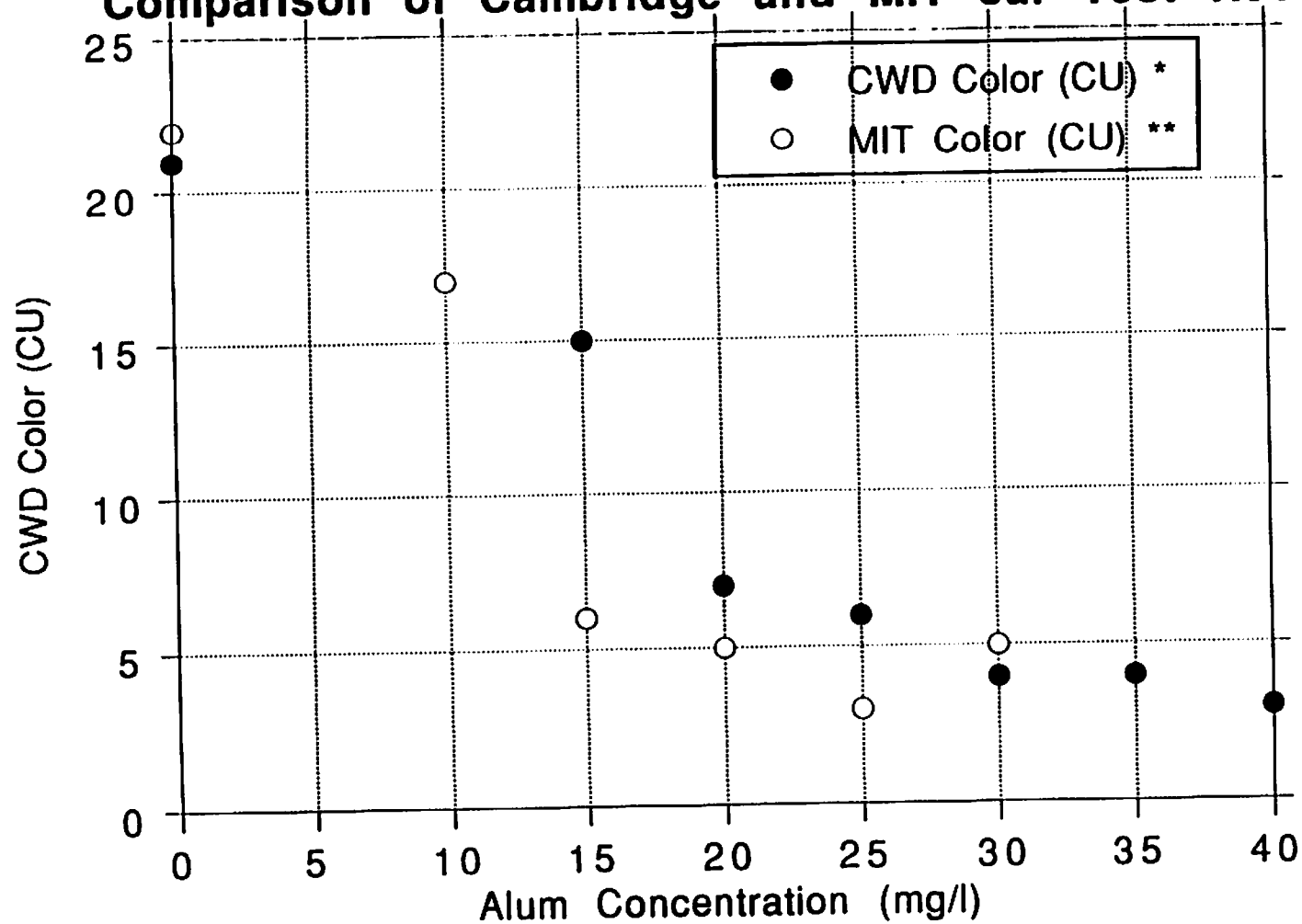
Figure 1b
Turbidity % Removal with Alum
Comparison of Cambridge and MIT Jar Test Results



* Average jar test data for Oct., Nov., and Dec., 1992

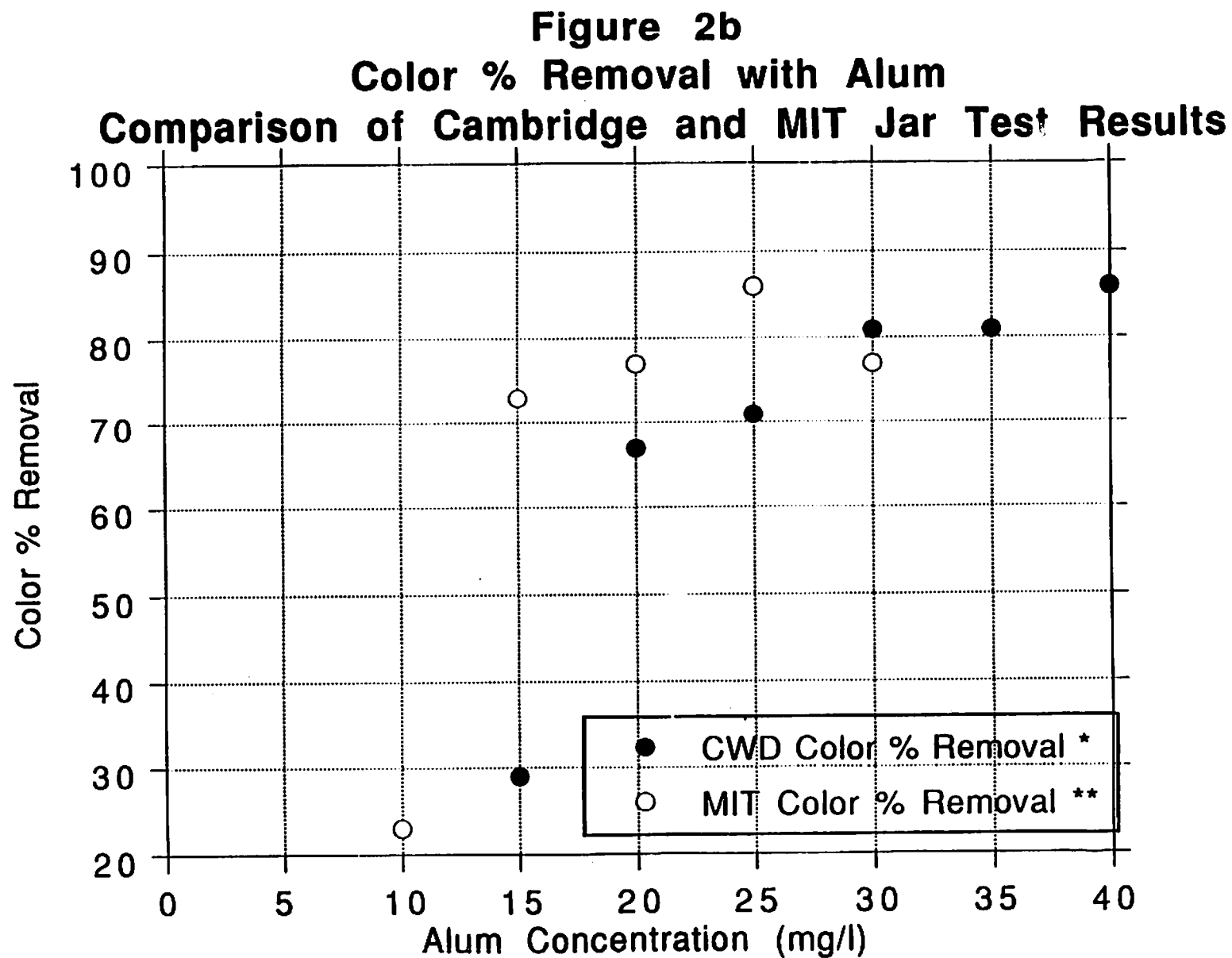
** Average jar test data for Nov. and Dec., 1992

Figure 2a
Color Concentration with Alum
Comparison of Cambridge and MIT Jar Test Results



* Average jar test results for Oct., Nov., and Dec., 1992

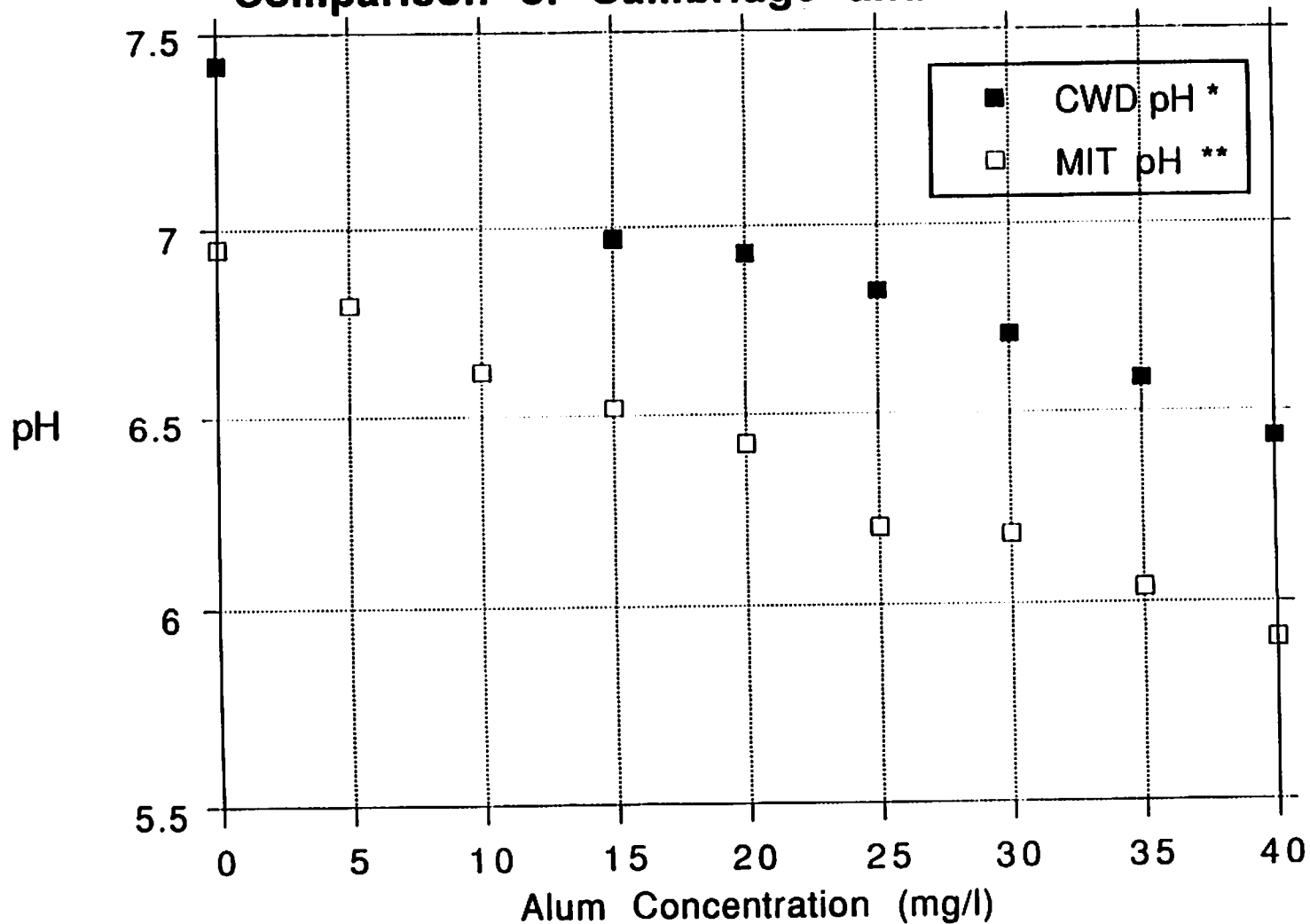
** Average jar test results for Nov. and Dec., 1992



* Average jar test data for Oct., Nov., and Dec., 1992

** Average jar test data for Nov. and Dec., 1992

Figure 3
pH vs. Alum Concentration
Comparison of Cambridge and MIT Jar Tests



* Average jar test data for Oct., Nov., and Dec., 1992

** Jar test data for 12/18/92

Figure 4a
Effect of Increased Bentonite Concentration on Turbidity

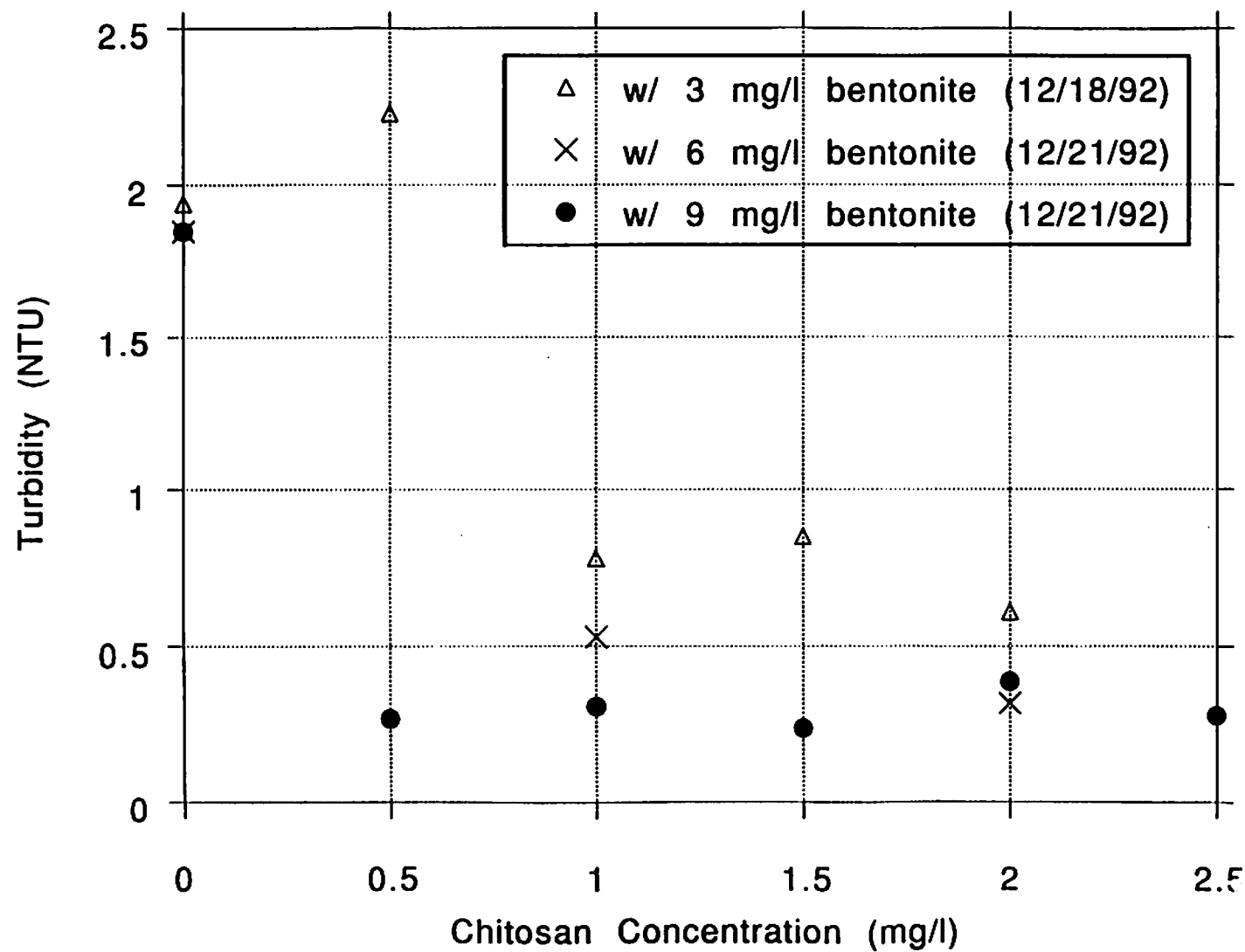


Figure 4b
Effect of Increased Bentonite Concentration on Color

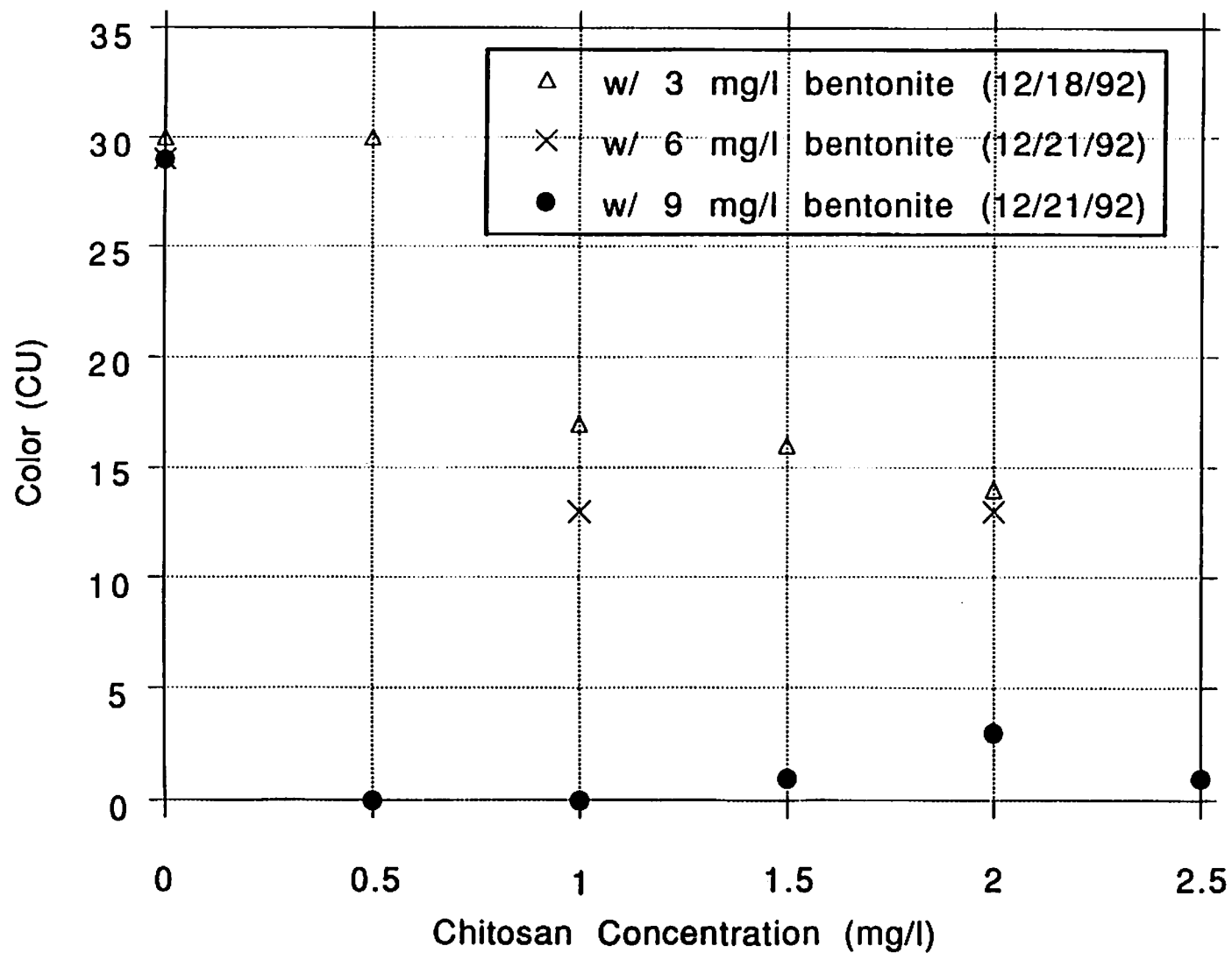
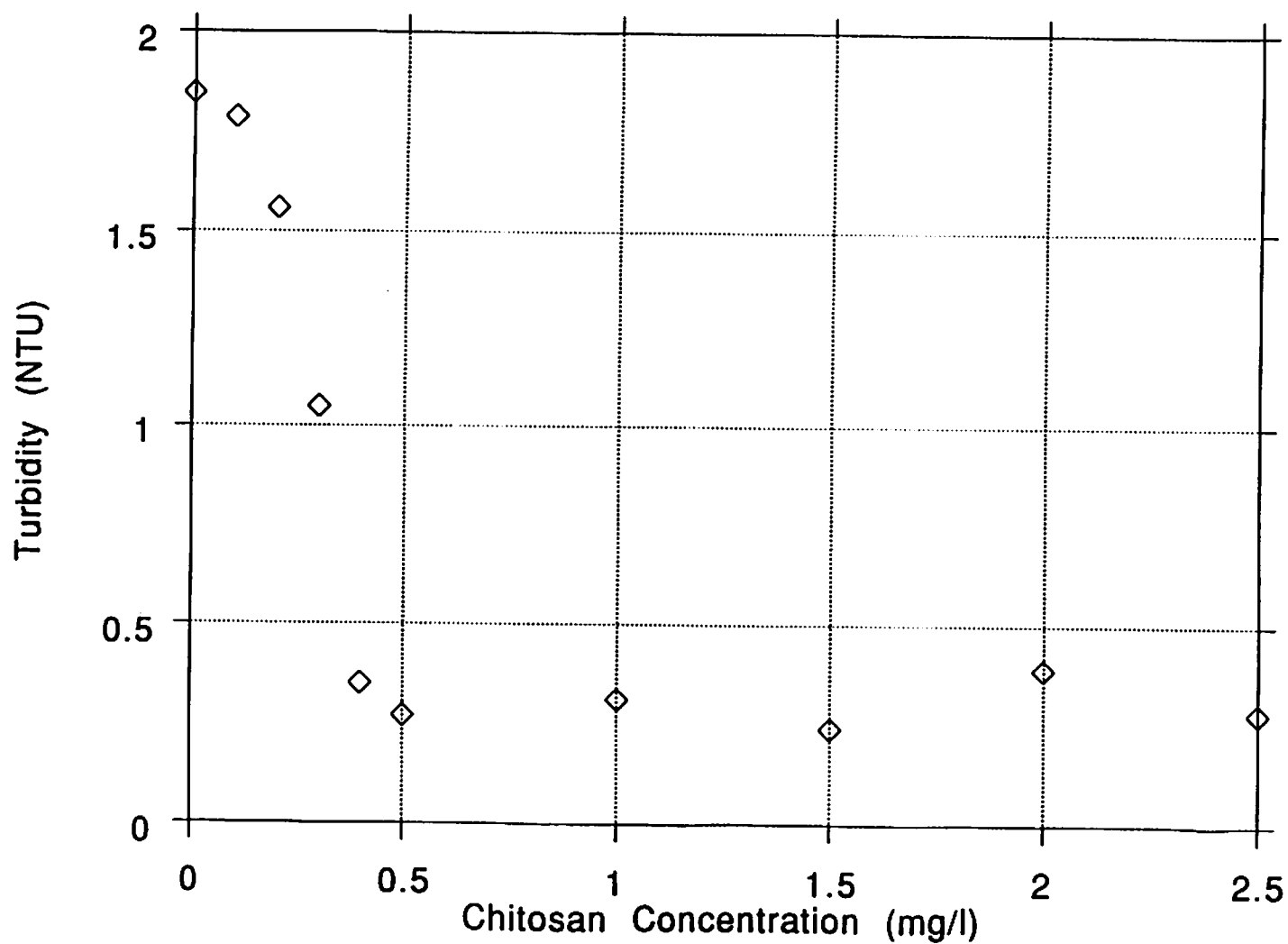


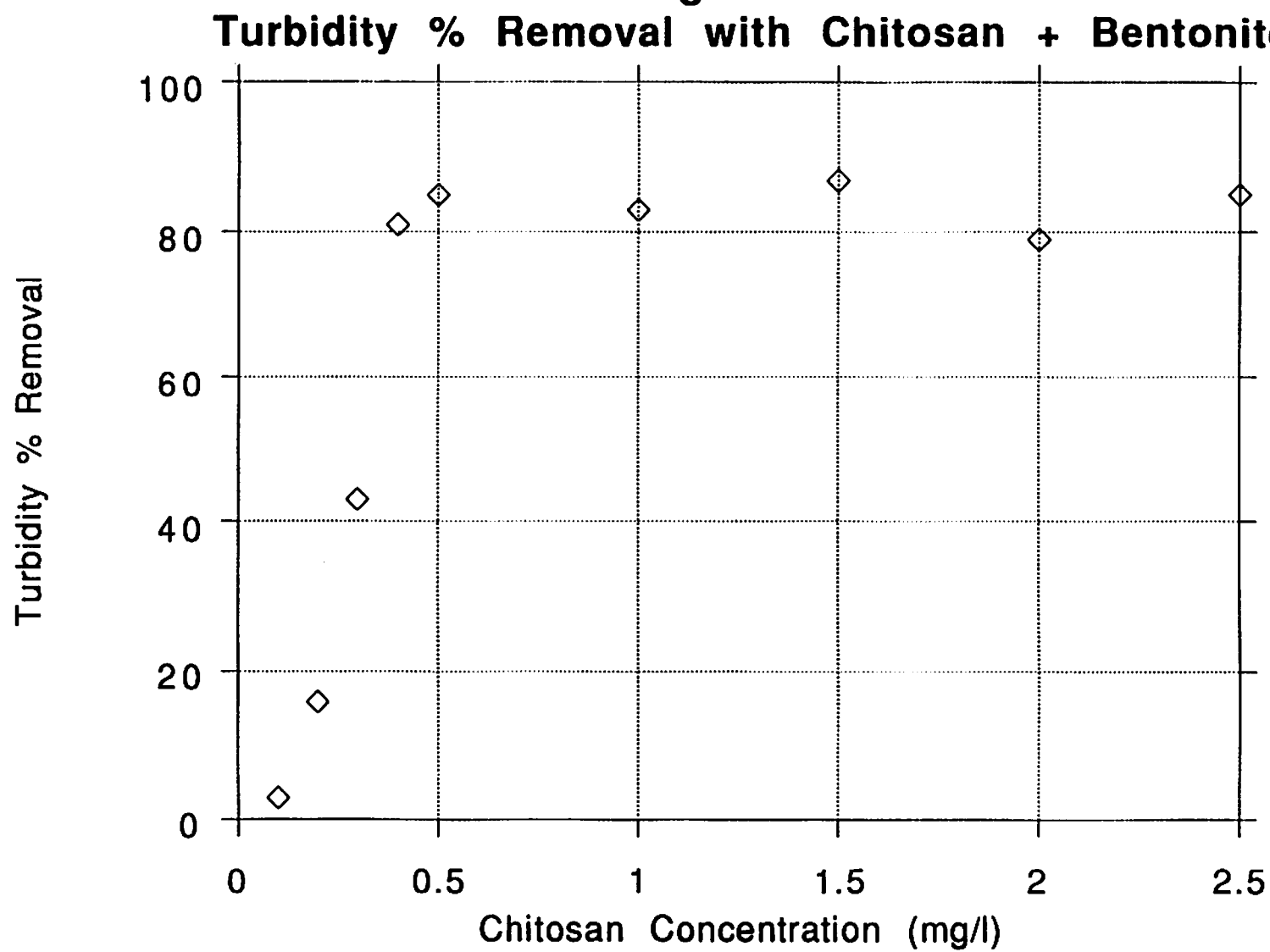
Figure 5a
Turbidity Concentration with Chitosan + Bentonite *



* Bentonite Concentration = 9 mg/l

(12/21/92)

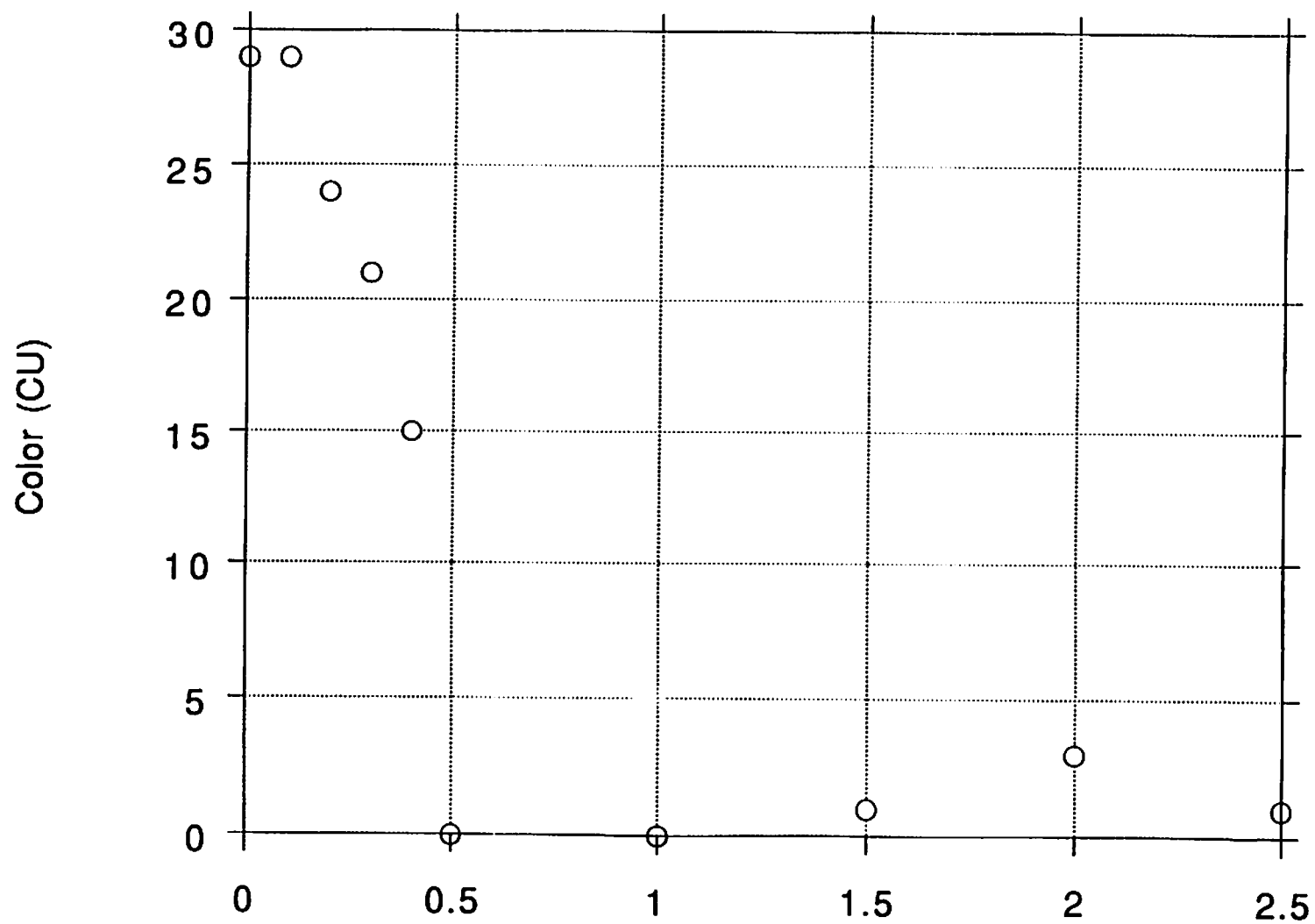
Figure 5b



* Bentonite Concentration = 9 mg/l

(12/21/92)

Figure 6a
Color Concentration with Chitosan + Bentonite *

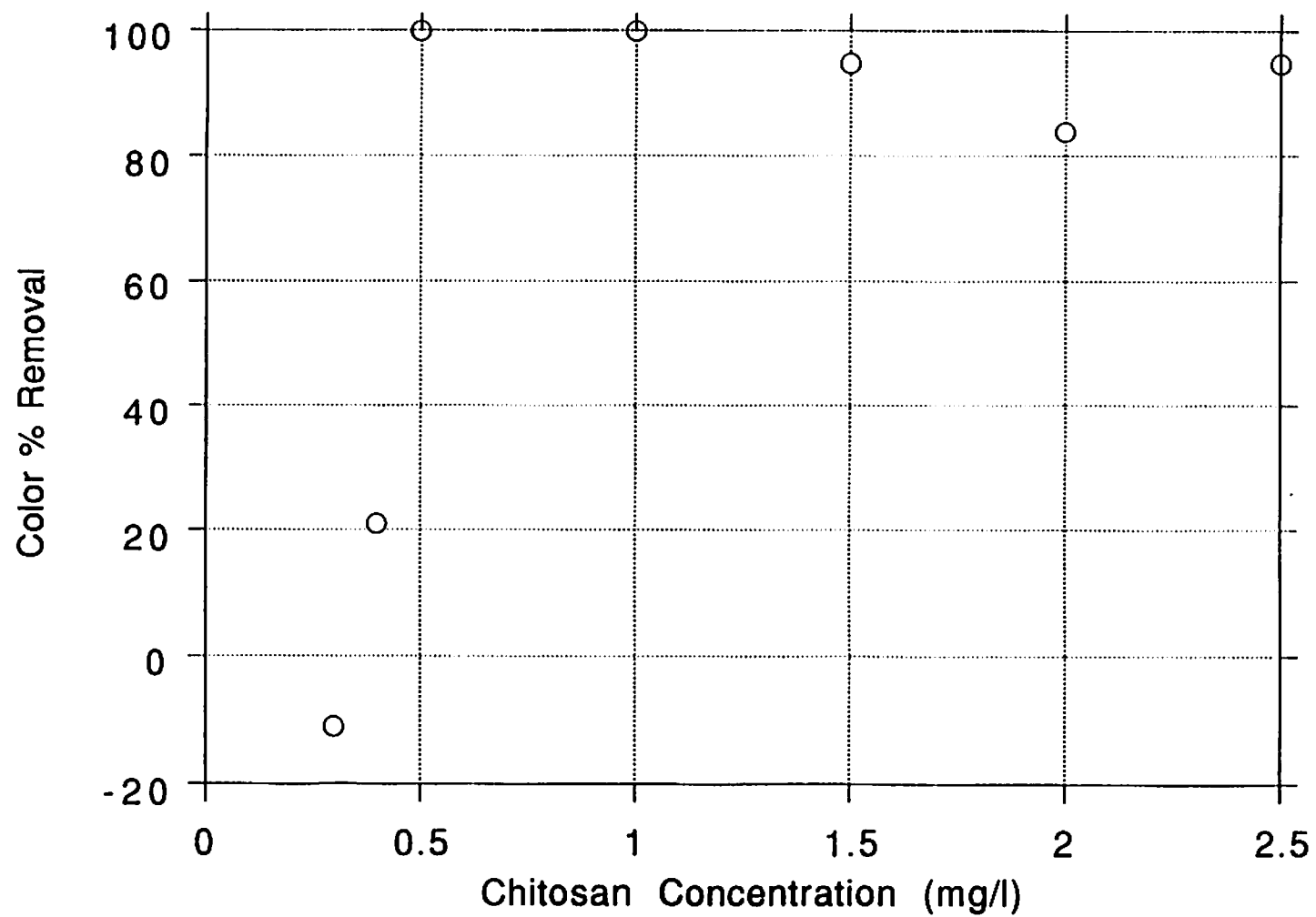


Chitosan Concentration (mg/l)
* Bentonite Concentration = 9 mg/l

(12/21/92)

Figure 6b

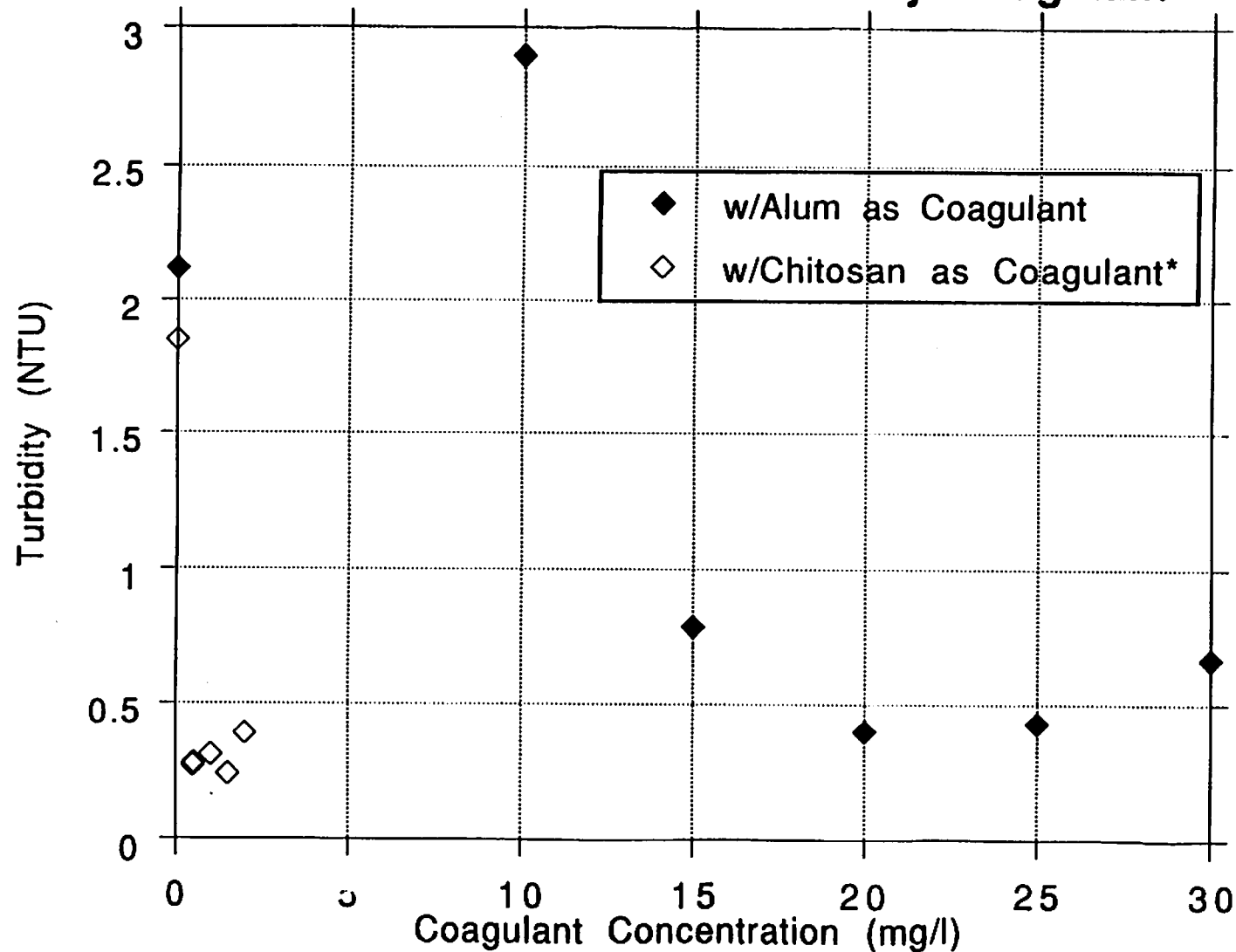
Color % Removal with Chitosan + Bentonite*



* Bentonite Concentration = 9 mg/l

(12/21/92)

Figure 7a
Turbidity Concentration Comparison
Alum or Chitosan as Primary Coagulant

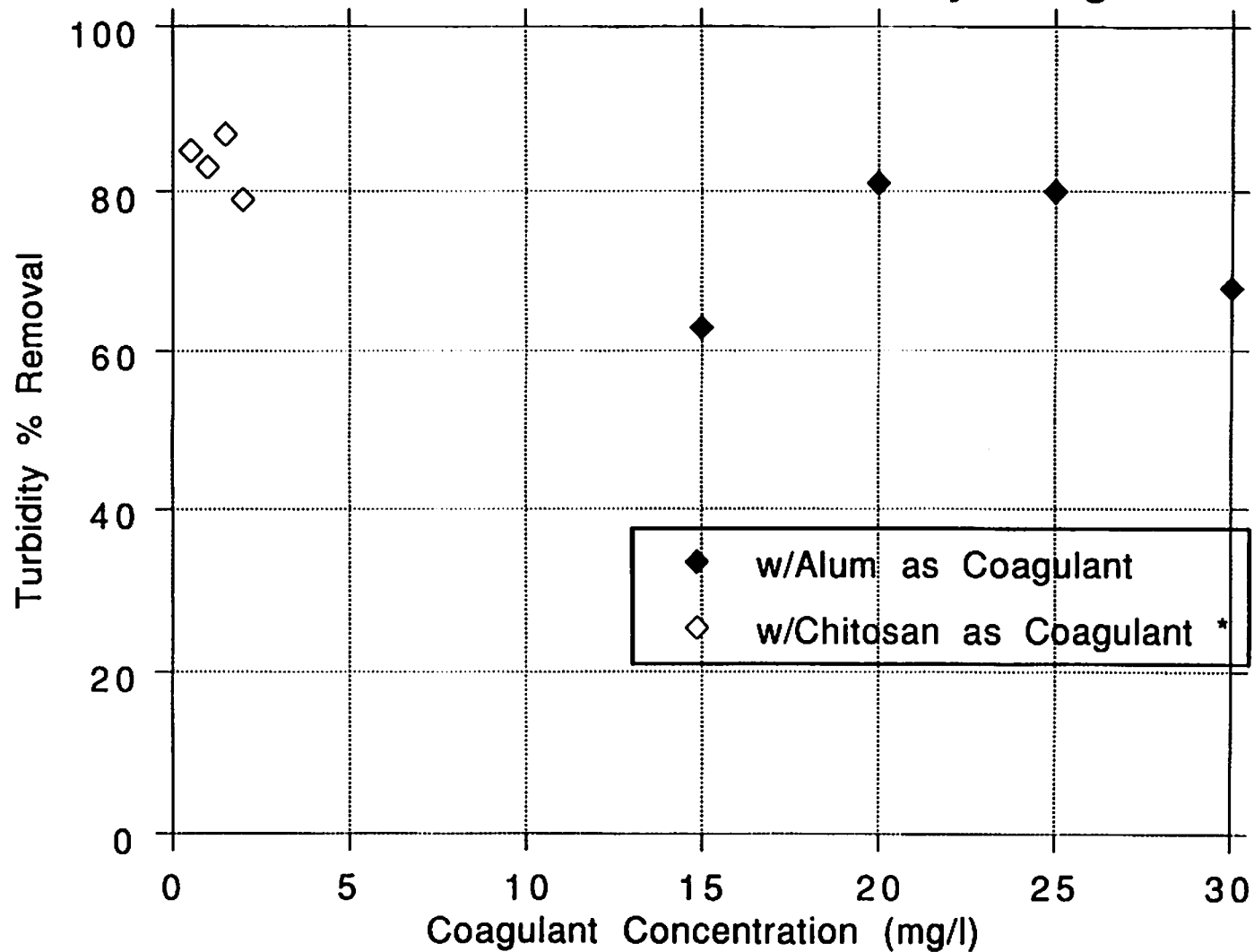


* 9 mg/l bentonite used as an aid with chitosan

Note: Coagulants were tested using their respective optimal mixing times and speeds

(12/21/92)

Figure 7b
Turbidity % Removal Comparison
Alum or Chitosan as the Primary Coagulant

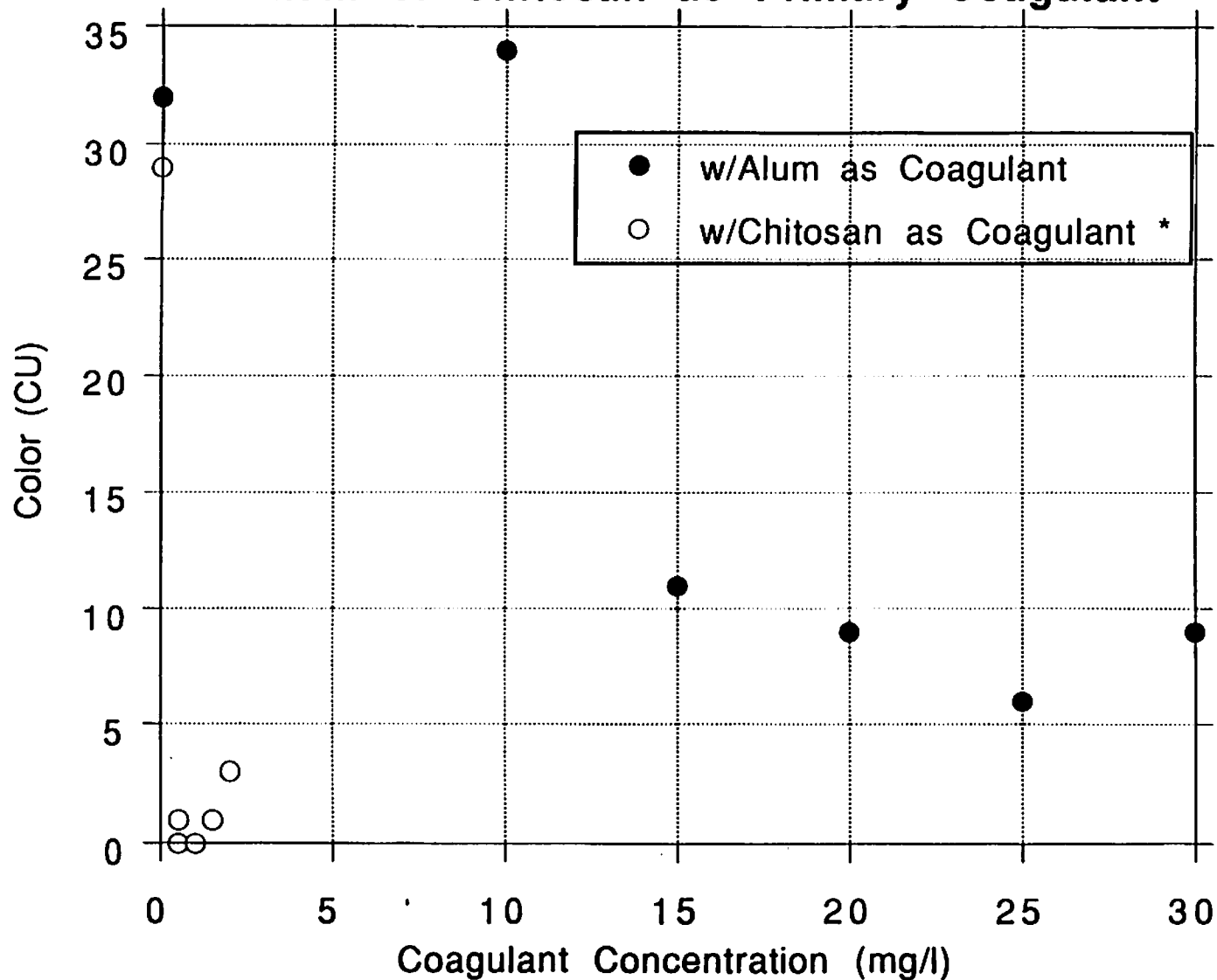


* 9 mg/l bentonite used as an aid with chitosan

Note: Coagulants were tested using their respective optimal mixing times and speeds

(12/21/92)

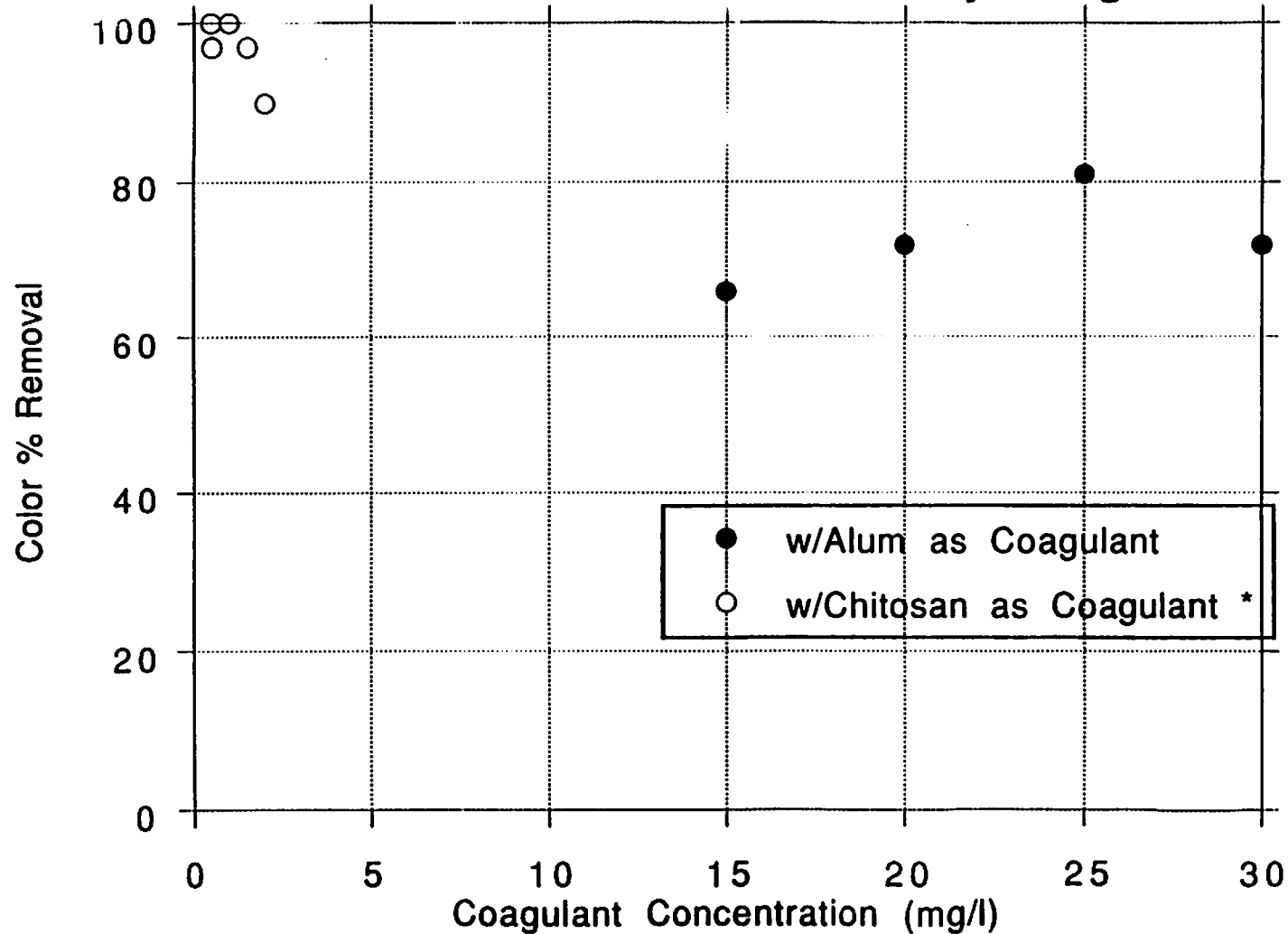
Figure 8a
Color Concentration Comparison
Alum or Chitosan as Primary Coagulant



* 9 mg/l bentonite used as an aid with chitosan

* Note: Coagulants were tested using their respective optimal mixing times and speeds (12/21/92)

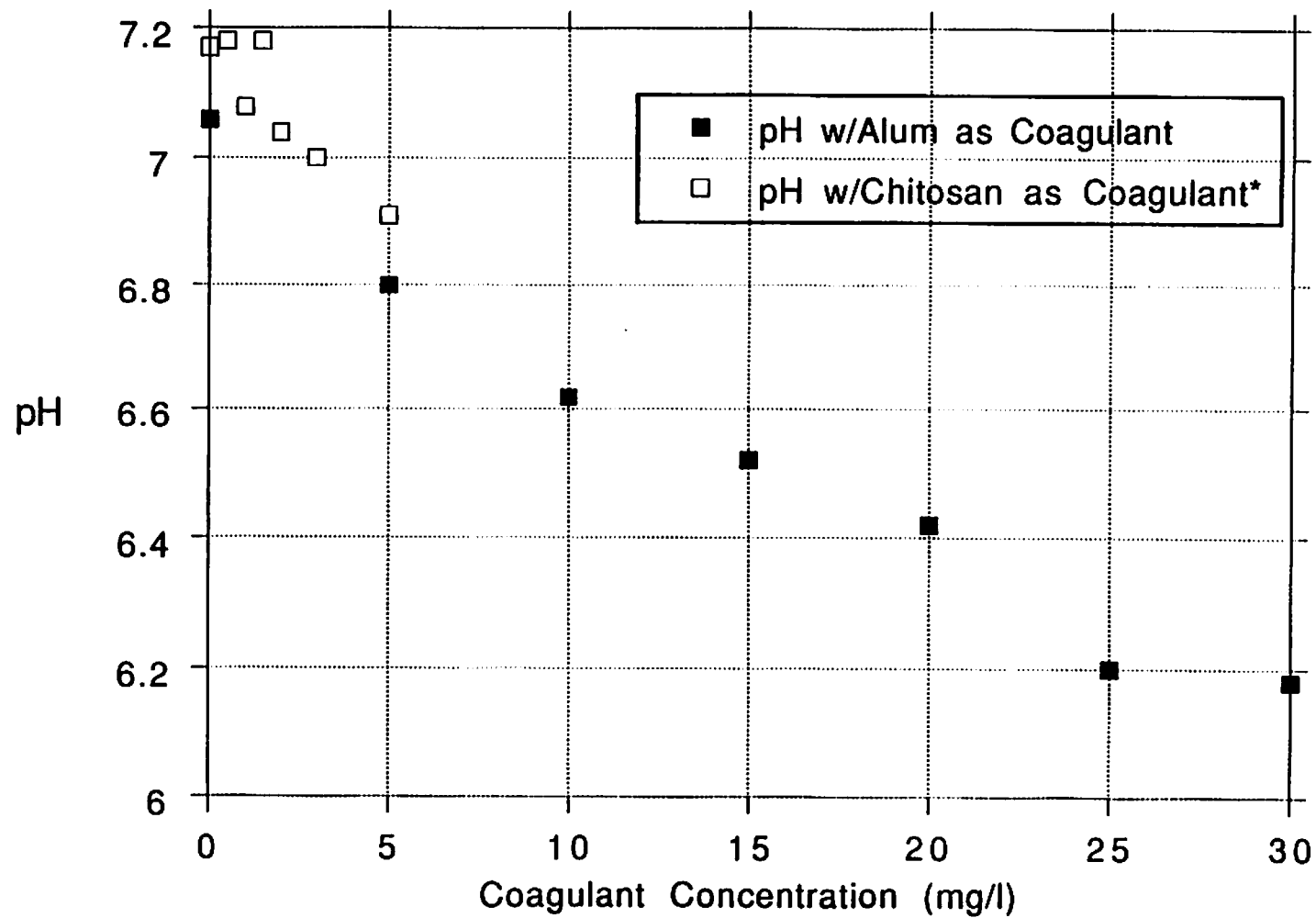
Figure 8b
Color % Removal Comparison
Alum or Chitosan as the Primary Coagulant



* 9 mg/l bentonite used as an aid with chitosan

Note: Coagulants were tested using their respective optimal mixing times and speeds (12/21/92)

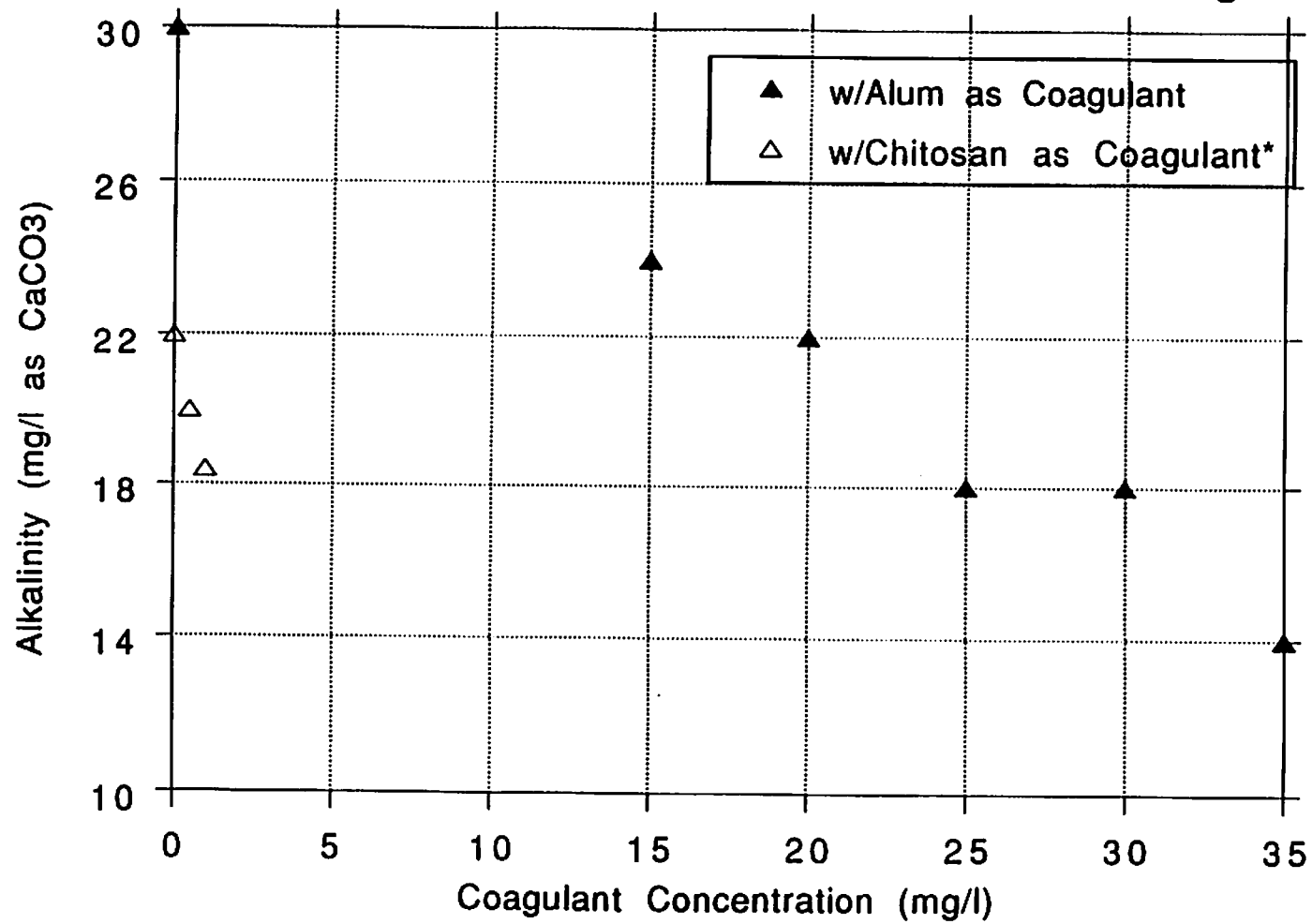
Figure 9
pH using Alum or Chitosan as the Coagulant



* 3 or 6 mg/l bentonite used as an aid with chitosan

(12/18/92)

Figure 10
Alkalinity using Alum or Chitosan as the Coagulant

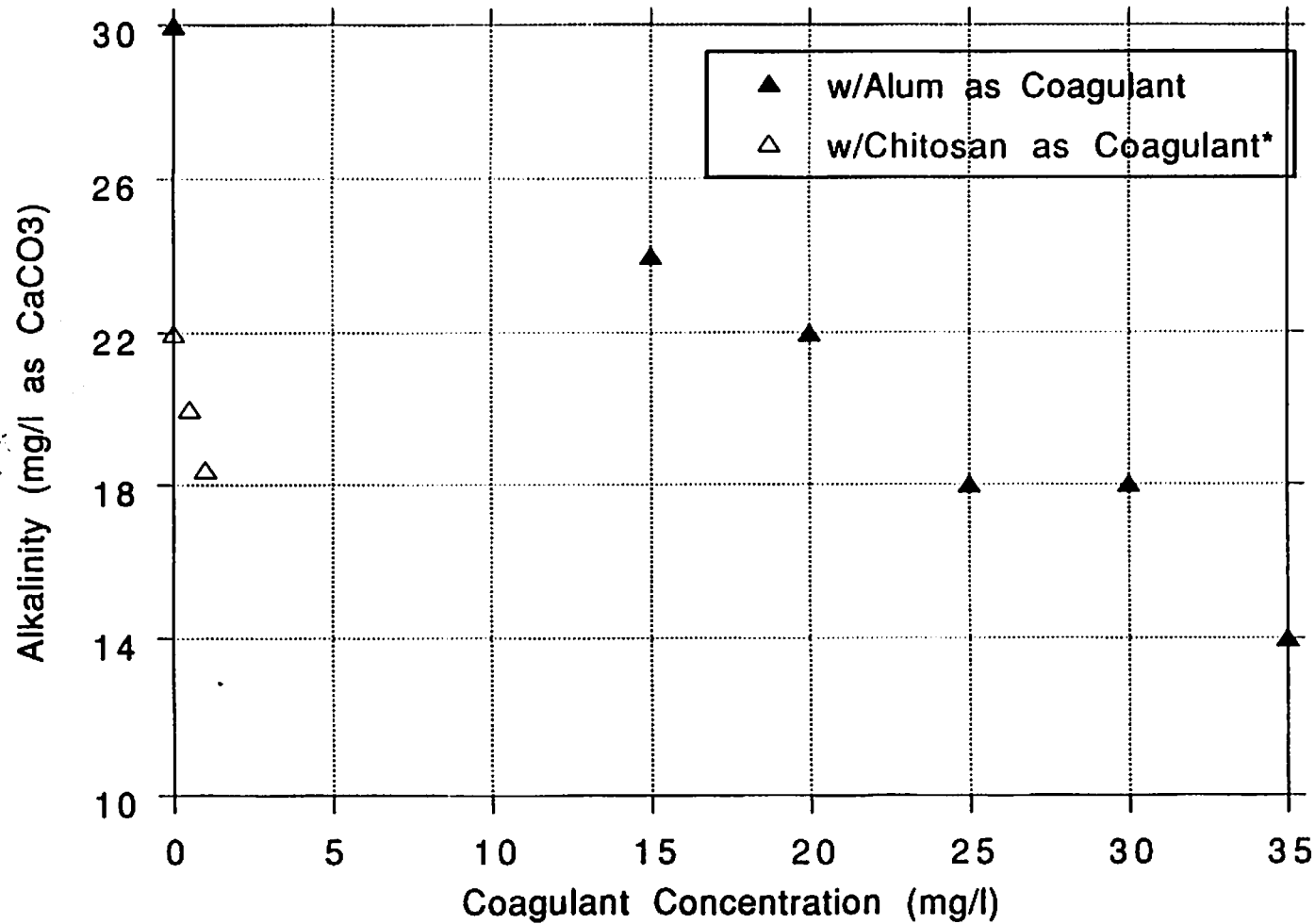


* 6 mg/l bentonite used as an aid with chitosan

(12/21/92)

Figure 10

Alkalinity using Alum or Chitosan as the Coagulant



* 6 mg/l bentonite used as an aid with chitosan

(12/21/92)